

U.S. DEPARTMENT OF COMMERCE, PATENT AND TRADEMARK OFFICE		DATE: December 28, 2000
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371		U.S. APPLICATION NO. (if known): 09/720645
INTERNATIONAL APPLICATION NO.: PCT/EP99/04560	INTERNATIONAL FILING DATE: JULY 1, 1999	PRIORITY DATE CLAIMED: JULY 2, 1998
TITLE OF INVENTION: PROCESS FOR THE PREPARATION OF AMMONIA		
APPLICANT(S) FOR DO/EO/US: Claus J. H. JACOBSEN and Michael BOE		
Applicant hereby submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:		
<ol style="list-style-type: none"> 1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371. 2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. 3. <input checked="" type="checkbox"/> This express request to begin national examination procedures (35 USC 371(f)) at any time rather than delay examination until the expiration of the time limit set in 35 USC 371(b) and PCT Articles 22 and 39(1). 4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date. 5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2)): <ol style="list-style-type: none"> a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau). b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau. c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US) 6. <input type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)). 7. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)) <ol style="list-style-type: none"> a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau). b. <input checked="" type="checkbox"/> have been transmitted by the International Bureau. c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. d. <input type="checkbox"/> have not been made and will not be made. 8. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). 9. <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). 10. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)). 		
ITEMS 11. TO 16. BELOW CONCERN OTHER DOCUMENT(S) OR INFORMATION INCLUDED:		
<ol style="list-style-type: none"> 11. <input checked="" type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98 together with the international search report and 6 references. 12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. ASSIGNEE NAME AND ADDRESS: <u>HALDOR TOPSØE A/S, Lyngby, Denmark</u> 13. <input checked="" type="checkbox"/> A FIRST preliminary amendment. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment 14. <input type="checkbox"/> A substitute specification. 15. <input type="checkbox"/> A change of power of attorney and/or address letter. 16. <input type="checkbox"/> Other items or information: 		

U.S. APPLICATION NO. (if known) 09/7 20645	INTERNATIONAL APPLICATION NO. PCT/EP99/04560	DATE: December 28, 2000	
--	--	--------------------------------	--

17. <input checked="" type="checkbox"/> The following fees are submitted: Basic National Fee (37 CFR 1.492(a)(1)-(5): Search Report has been prepared by the EPO or JPO: \$860.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) \$690.00 No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)) \$710.00 Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$1000.00 International preliminary examination fee (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4) \$100.00 <div style="text-align: right;">ENTER APPROPRIATE BASIC FEE AMOUNT = \$ 860.00</div>	<u>CALCULATIONS</u>	<u>PTO USE ONLY</u>
---	---------------------	---------------------

Surcharge of \$130.00 for furnishing the oath or declaration later than __ 20 x 30 months from the earliest claimed priority date (37 CFR 1.492(e)).	\$ 130.00	
--	------------------	--

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
TOTAL	2 -20 =		X \$ 18.00		
INDEPENDENT	1- 3 =		X \$ 80.00		
Multiple dependent claims(s) (if applicable)			+ \$270.00		
TOTAL OF ABOVE CALCULATIONS =				\$ 990.00	
Reduction by 1/2 for filing by small entity, if applicable. (Note 37 CFR 1.9, 1.27, 1.28).					
SUBTOTAL =				\$ 990.00	
Processing fee of \$130.00 for furnishing the English translation later than __ 20 __ 30 months from the earliest claimed priority date (37 CFR 1.492(f)). +					
TOTAL NATIONAL FEE =				\$ 990.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +					
TOTAL FEES ENCLOSED =				\$ 990.00	
				Amount to be:	
				refunded	\$ _____
				charged	\$ _____
					\$ _____

ATTORNEY'S DOCKET NO: 001703

U.S. APPLICATION NO.
(if known)

09/720645

INTERNATIONAL APPLICATION NO.

PCT/EP99/04560

DATE: December 28, 2000

- a. ☒ A check in the amount of \$ **990.00** to cover the above fees is enclosed. (\$860.00 for filing fee and \$130.00 for late filing of the declaration). (This paper is filed in triplicate)
- b. ☐ Please charge my Deposit Account No. 01-2340 in the amount of \$___ to cover the above fees. (A duplicate copy of this sheet is enclosed.)
- c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 01-2340.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed to request that the application be restored to pending status.

Send All Correspondence To:



23850

PATENT TRADEMARK OFFICE

SIGNATURE

Le-Nhung McLeland

NAME

31,541

REGISTRATION NUMBER

NM/yap

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Attached Patent Application of:

Claus J. H. JACOBSEN et al.

Serial No.: (to be assigned)

Group Art Unit: (to be assigned)

Filed: HEREWITH

Examiner: (to be assigned)

For: PROCESS FOR THE PREPARATION OF AMMONIA

PRELIMINARY AMENDMENT

Director of Patents and Trademarks
Washington, D.C. 20231

Date: December 28, 2000

Sir:

Prior to calculation of the filing fee and examination of this application, please amend the above-identified application as follows:

IN THE CLAIMS:

Please amend the claims as follows:

Please cancel claims 1-4, and add the following new claims 5 and 6.

-5. Process for the preparation of ammonia by contacting an ammonia synthesis gas with ammonia catalyst particles arranged in a fixed bed comprising catalyst particles with a particle size in the range of ≥ 0.2 mm to < 1.5 mm, said synthesis gas being passed in radial direction through said fixed bed, wherein said fixed bed contains a mixture of catalyst particles with a size of 1.5 - 3.0 mm, 0.8 - 1.5 mm and 0.3 - 0.8 mm in a volume ratio of (40 - 70):(10 - 40):(10 - 30).--

-6. The process of claim 5, wherein the fixed bed contains at least 10% by volume of catalyst particles having a particle size in the range of ≥ 0.2 mm to < 1.5 mm.--

REMARKS

Claim 5 is a combination of original claims 1 and 3. Claim 6 recites the same additional limitation as in original claim 2.

In the event that any fees are due in connection with this paper, please charge our Deposit Account No. 01-2340.

Respectfully submitted,

ARMSTRONG, WESTERMAN, HATTORI,
McLELAND & NAUGHTON



Le-Nhung McLeland
Attorney for Applicants
Reg. No. 31,541

Atty. Docket No.: 001703

1725 K Street, N.W., Suite 1000
Washington, DC 20006

Tel: (202) 659-2930

Fax: (202) 887-0357

LNM:ann

- 1 -

Process for the Preparation of Ammonia

The present invention is directed to the preparation of ammonia by catalytic conversion of an ammonia synthesis gas.

5 Conventionally, industrial ammonia synthesis is based on conversion of ammonia synthesis gas consisting of hydrogen and nitrogen in a substantially stoichiometric mole ratio of 3:1. The synthesis gas is passed at high pressure through a fixed bed of ammonia catalyst particles of mainly
10 magnetite, which is converted by reduction into the catalytically active form of α -iron.

The process performance is governed not only by the catalyst composition, but also by the size and shape of the
15 catalyst particles. For ammonia synthesis processes operating at catalyst beds with an axial synthesis gas flow the usual catalyst particle size is in the range of 6-10 mm.

Due to a reduced flow resistance in radial flow type ammonia reactors the catalyst particle size employed in these
20 reactors is between 1.5 and 3 mm.

It has now been found that process performance of ammonia synthesis still may be improved in terms of a higher ammonia product yield when employing in radial ammonia reactors
25 a fixed catalyst bed of ammonia catalyst with a mixed composition of catalyst particles having a large size and small size. A mixture of large size and small size particles results in higher bulk density due to smaller particles pack in voids being formed between larger particles.
30 Higher bulk density provides an increased amount of catalyst in the ammonia reactor resulting in a higher catalytic activity per reactor volume.

- 2 -

Pursuant to the above finding, this invention is a process for the preparation of ammonia by contacting an ammonia synthesis gas with ammonia catalyst particles arranged in a fixed bed, wherein the fixed bed comprises catalyst particles of the ammonia catalyst with a particle size being in the range of less than 1.5 mm and larger than or equal with 0.2 mm.

By inclusion of a significant amount of particles with a size within the specified range, the bulk density increases causing a higher pressure drop over the catalyst bed, and, thereby, an improved flow distribution of the synthesis gas within the bed.

When operating the inventive process at industrial conditions an improved flow distribution of synthesis gas is obtained when the catalyst bed contains between 10% and 80% by volume of ammonia catalyst particles having a particle size below 1.5 mm.

The Table below summarizes the relative density of different particle sizes of conventional ammonia catalysts commercially available from Haldor Topsøe A/S.

Table

Particle Size/mm		ρ rel.
1.5-3.00		1.00
0.8-1.5		0.97
0.3-0.8		0.95
60%	1.5-3.0 +	1.09
20%	0.8-1.5 +	
20%	0.3-0.8	

- 3 -

A mixture containing 60%, 20% and 20% of 1.5-3 mm, 0.8-1.6 mm and 0.3-0.8 mm particles, respectively, has a relative bulk density of 1.09.

- 5 The absolute bulk density of the industrial catalyst depends on the loading procedure, however, the same relative density can be found.

10 Inclusion of 0.2-1.5 mm sized catalyst particles provides higher catalyst bulk density, and also a lower diffusion resistance. By the broader particle size distribution and the increased bulk density a higher pressure drop is obtained over the catalyst bed causing a significant improved flow distribution of the synthesis gas in the catalyst bed.

15

At present a preferred particle size distribution of ammonia catalyst arranged as fixed bed is obtained by mixing particles with a size of 1.5-3.0 mm, 0.8-1.5 mm, and 0.3-0.8 in a weight ratio of 40-70 : 10-40 : 10-30.

20

- 4 -

CLAIMS

1. Process for the preparation of ammonia by contact-
ing an ammonia synthesis gas with ammonia catalyst par-
5 ticles arranged in a fixed bed, wherein the fixed bed
comprises catalyst particles of the ammonia catalyst with a
particle size being in the range of <1.5 mm and ≥ 0.2 mm.

2. The process of claim 1, wherein the fixed bed
10 contains at least 10% by volume of catalyst particles
having a particle size in the range of <1.5 mm and ≥ 0.2 mm.

3. The process of claim 1, wherein the fixed bed
contains a mixture of particles with a size of 1.5-3.0 mm,
15 0.8-1.5 mm and 0.3-0.8 mm in a volume ratio of
40-70:10-40:10-30.

4. The process according to anyone of the preceding
claims, wherein the synthesis gas is passed in radial
20 direction through the fixed bed.

Declaration for U.S. Patent Application

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention **entitled** (Insert Title) PROCESS FOR THE PREPARATION OF AMMONIA the specification of which is attached hereto unless the following is checked

☒ was filed on July 1, 1999 as PCT International Application Number PCT/EP99/04560 and was amended on _____ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claim(s), as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, § 119 (a) - (d) of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application for which priority is claimed:

(List prior foreign applications. See note A on back of this page)	<u>PA 1998 00892</u> (Number)	<u>Denmark</u> (Country)	<u>2/July/1998</u> (Day/Month/Year Filed)	Priority Claimed <u>XX</u> Yes ___ No
	_____ (Number)	_____ (Country)	_____ (Day/Month/Year Filed)	___ Yes ___ No
	_____ (Number)	_____ (Country)	_____ (Day/Month/Year Filed)	___ Yes ___ No
	_____ (Number)	_____ (Country)	_____ (Day/Month/Year Filed)	___ Yes ___ No
	_____ (Number)	_____ (Country)	_____ (Day/Month/Year Filed)	___ Yes ___ No

(See note B on back of this page) ___ See attached list for additional prior foreign applications

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

(List Prior U.S. Applications)	<u> </u> (Appln. Serial No.)	<u> </u> (Filing Date)	<u> </u> (Status: Patented, Pending, Abandoned)
	<u> </u> (Appln. Serial No.)	<u> </u> (Filing Date)	<u> </u> (Status: Patented, Pending, Abandoned)
	<u> </u> (Appln. Serial No.)	<u> </u> (Filing Date)	<u> </u> (Status: Patented, Pending, Abandoned)

PA 32476-0007/612

I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:



23850

PATENT TRADEMARK OFFICE

Please direct all communications to the following address:



23850

PATENT TRADEMARK OFFICE

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Title 18 of the United States Code, § 1001 and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

(See note C above)

1-00
Full name of sole or first inventor (given name, family name) Claus J.H. JACOBSEN

Inventor's signature *Claus J.H. Jacobsen* Date 10/6-2001

Residence Jaegerspris, DENMARK DKX Citizenship Danish

Post Office Address Runestien 9 DK-3630 Jaegerspris DENMARK

2-00
Full name of second inventor (given name, family name) Michael BOE

Inventor's signature *Michael Boe* Date 12/6-2001

Residence Klampenborg, DENMARK DKX Citizenship Danish

Post Office Address Taarbaek Strandvej 86, St. DK-2930 Klampenborg DENMARK

Full name of third inventor (given name, family name) _____

Inventor's signature _____ Date _____

Residence _____ Citizenship _____

Post Office Address _____

Full name of fourth inventor (given name, family name) _____

Inventor's signature _____ Date _____

Residence _____ Citizenship _____

Post Office Address _____

Full name of fifth inventor (given name, family name) _____

Inventor's signature _____ Date _____

Residence _____ Citizenship _____

Post Office Address _____